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Ship Materials Engineering Department

Research and Development Report

Development of Neodymium and Er_3Ni Regenerator Materials

by

Louis F. Aprigliano, Geoffrey Green, James Chafe,
Lisa O'Connor, Frank Biancanello* and Steve Ridder*

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ABSTRACT

Neodymium and Er_3Ni powders were produced and examined as possible candidates for use as regenerator matrices in the regenerative heat exchanger of the Gifford-McMahon cycle refrigerator. In the case of Er_3Ni, crushed powders were brittle and angular in shape, molten salt produced spheres were heavily oxidized, and gas atomized powder had a low yield (3 %) and a large fraction of hollow particles. Neodymium powder, which was produced by the rotating electrode method, was smooth and spherical in shape and had a high yield (15 %) of solid particles.

ADMINISTRATIVE INFORMATION

This report is submitted in fulfillment of Milestone 1, Task 1, of the Supeconducting Propulsion Project (RH21E46) of the Surface Ship Technology Block Program (ND1A/PE062121N). The work was sponsored by the Office of Naval Technology (ONT 211) and performed by the David Taylor Research Center (DTRC 2812 and 2712).

INTRODUCTION

The Gifford-McMahon (GM) refrigeration cycle can be used to meet the needs of many systems that require cryogenic cooling. The GM cycle makes use of the regenerative heat exchanger. A crucial component of the regenerative heat exchanger is the regenerative matrix. The function of the regenerative matrix is depicted in

Figure 1. A typical regenerative matrix consists of lead shot measuring approximately 0.23 mm (0.009 inch) in diameter.

The heat capacity of the regenerative matrix is an indicator of its performance. A plot of heat capacity versus temperature for lead is shown in Figure 2[1]. In the case of lead, the drop-off in the heat capacity below 12 K is a significant factor that limits its effective cooling to 10 K. The heat capacities of the candidate regenerative matrices, neodymium[2] (Nd) and erbium-3-nickel[3] (Er_3Ni), are shown in Figure 2. Both show a favorable heat capacity spike at approximately 6 K and both retain some heat capacity to 4 K. This feature makes them an attractive alternative to lead in the temperature range of 4 to 10 K.

Making powders of Nd and Er_3Ni is not a trivial process. Both are highly reactive with oxygen which makes them difficult to produce as quality powders. Furthermore, the Er_3Ni is extremely brittle and there is some concern that the long term operation in the regenerative heat exchanger of a GM cycle can break the powder down into very fine particles. These fine particles can escape from the regenerative heat exchanger and adversely affect other GM cooler components (e.g. the displacer, seals, and valves).

The purpose of this study is to evaluate the quality of various sources of Er_3Ni and Nd powders. This is being done because of the above mentioned features in the heat capacity of these two materials and because regenerative matrices made with powdered Er_3Ni have been reported[3,4,5] to improve the performance of the GM cycle. However, the work with Er_3Ni has provided very little information on how the powders were produced, on the process yields or on the quality. In addition to addressing these issues, we have studied the performance of these powders in a GM cooler. Those results have been reported[6] separately.

EXPERIMENTAL PROCEDURE

Er_3Ni

Commercial sources were used to obtain Er_3Ni in the form of crushed powder and molten salt bath produced spheres. In addition, the Gaithersburg branch of the National Institute of Standards and Technology (NIST) produced spherical powders of Er_3Ni using an inert gas atomization process. The crushed powder is made by breaking up a solid bar of Er_3Ni until powders that will pass a 0.25 mm (0.010 inch) sieve are produced. The molten salt bath involves melting a supply of Er_3Ni nuggets and allowing the molten particles to slowly settle and drift in a column of molten salt. Inert gas atomization involves evacuating a large cylindrical chamber -- 1.6 m (46 inch) inside diameter by 2.7 m (9 feet tall) -- and then backfilling to one atmosphere of argon. At the top is a melt chamber with an induction melting crucible supported on a stainless steel separating plate. A stopper rod is used to contain the liquid metal in the crucible. When the stopper rod is raised the molten metal flows through a ceramic delivery tube. The liquid forms a sheet at the nozzle tip which is sheared by the atomizing gas into first ligaments, then dumbbells and finally droplets. The droplets fall in the main chamber and solidify into spherical powders which are collected at the base in a cyclone separator.

NEODYMIUM

A commercial source was used to obtain Nd in the form of spherical powders by the method of rotating plasma electrode (RPE). For this procedure, a bar of Nd, 6.35 cm (2.5 inches) in diameter and 25.4 cm (10 inches) long, is machined from a commercially supplied ingot of Nd. The bar is rotated along its long axis at high speed in a chamber filled with argon. An electrode is brought close to the end of the Nd bar and an electrical arc is struck. The combination of the heating and the rotational forces causes molten particles of Nd to be spun off and to solidify into spherical powders.

(An attempt has not been made to make Er_3Ni by the RPE method. Er_3Ni is very brittle and would be difficult to machine with the dimensional tolerances needed for the electrode in the RPE method.)

EXAMINATION

Optical and electron microscopy were utilized to examine the condition of the powders. The powders were embedded in epoxy and then in bakelite for polishing to a flat surface. This allowed the interior of the particles to be examined for voids and allowed for the use of the electron microprobe to study the composition and microstructure of the powders. The scanning electron microscope was used to study the surface topography of the powders and to measure their size.

RESULTS

Er_3Ni

Scanning electron and electron microprobe examination showed the crushed powder to be very angular in shape (Figure 3) and the molten salt powder to be coated with a heavy oxide layer (Figure 4). Neither features are considered desirable in a good regenerator matrix. The spray atomized powder was spherical and was free of a heavy oxide, but the insides of many of the spheres were hollow (Figure 5). An x-ray diffraction analysis of this powder showed it to be predominately Er_3Ni with a minor amount of Er_2O_3.

For the following reasons, hollow spheres are not preferred for use in a regenerator matrix. A powder size of 0.23 mm (0.009 inch) diameter is considered optimum for a regenerator matrix in a regenerative heat exchanger. During each cycle the regenerator matrix must alternately transfer heat to and from the helium that is compressed and expanded in the GM cooling cycle. If the powder particles in the regenerator matrix are too small or too large, the particle mass is not being used effectively. Likewise, if the particles are hollow, the mass needed to optimize the heat flow during each cycle is no longer present.

The inert gas atomized powder had a low yield (approximately 3 %) in the size range of 0.12 to 0.25 mm (0.005 to 0.010 inch). Most of the particles are much smaller than this. As currently designed, the NIST gas atomizer favors the production of finely size particles. However, it can be modified to produce a higher yield of larger particles. Such a modification is being planned.

NEODYMIUM

The Nd powder was visually bright and shiny and could be handled in room air for brief periods without violent or excessive oxidation. Scanning electron microscope examination showed the Nd powder to be very smooth in surface texture and to have a high proportion of spherical particles (Figure 6). Electron microprobe examination showed that the particles did not have an excessively thick oxide layer and were solid throughout their cross-section. The yield (approximately 15 %) was acceptable in the size range 0.12 to 0.25 mm (0.005 to 0.010 inch). Since this yield was obtained on the first run, we believe that continuing efforts will further improve the yield.

CONCLUSION

1. Spherical Nd powder in the size range of 0.12 to 0.25 mm (0.005 to 0.010 inch) can be made with high yields (approximately 15 %) by the rotating plasma electrode method.

2. Due to the combined brittle and highly reactive nature of Er_3Ni, only the inert gas atomization process was able to produce spherical powder without excessive oxidation. However, more process experimentation and system modifications are needed if the yield of solid particles with sizes greater than 0.25 mm (0.005 inch) is to be increased.

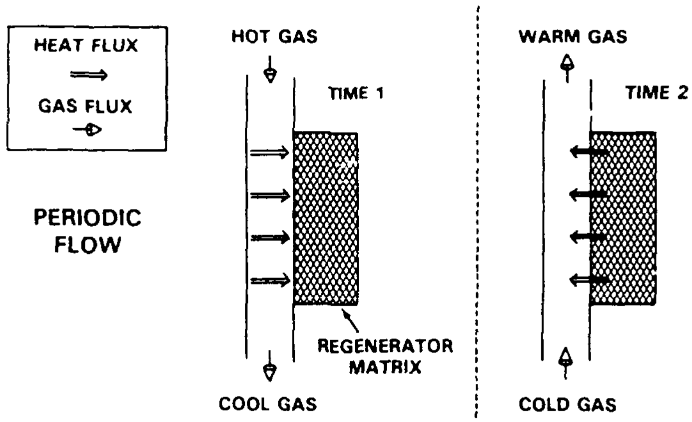


Fig. 1. Schematic representation of the function of a regenerator matrix in a regenerative heat exchanger.

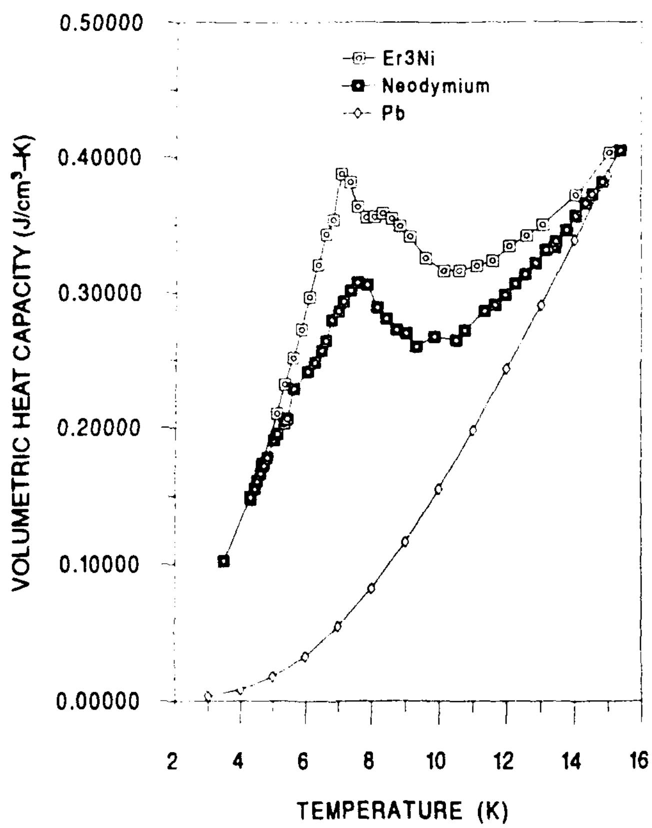


Fig. 2. Heat capacity versus temperature for lead, neodymium and Er_3Ni.

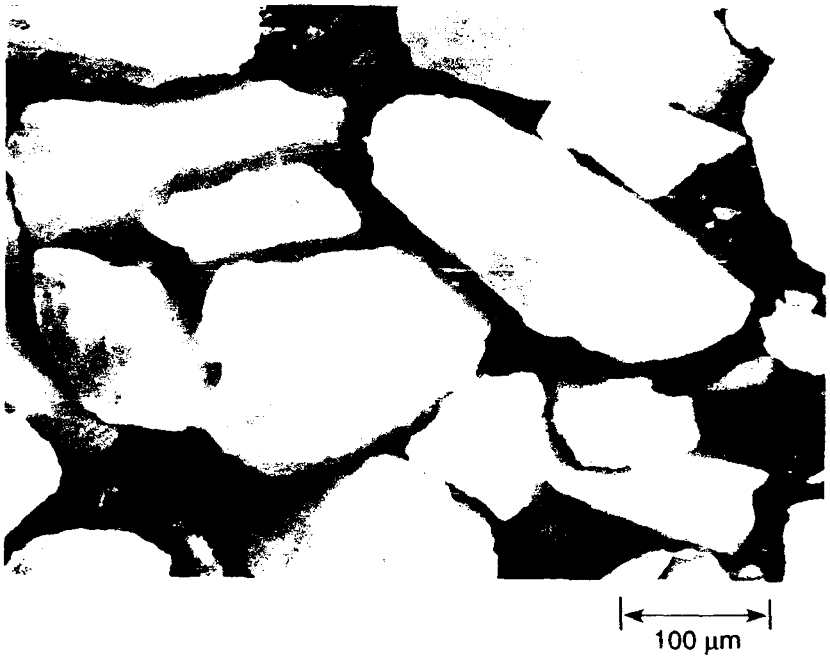


Fig. 3. Er_3Ni crushed powder; scanning electron micrograph.

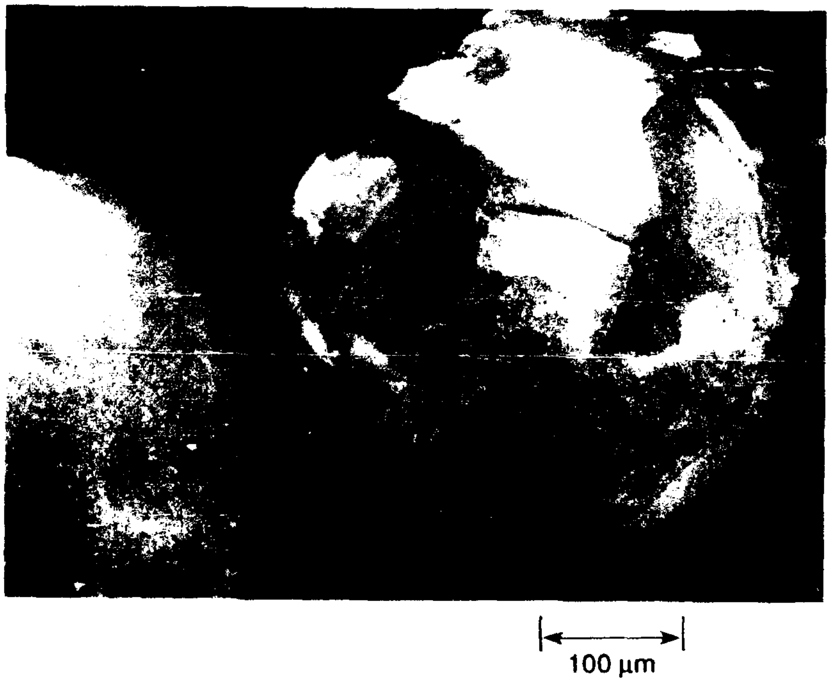
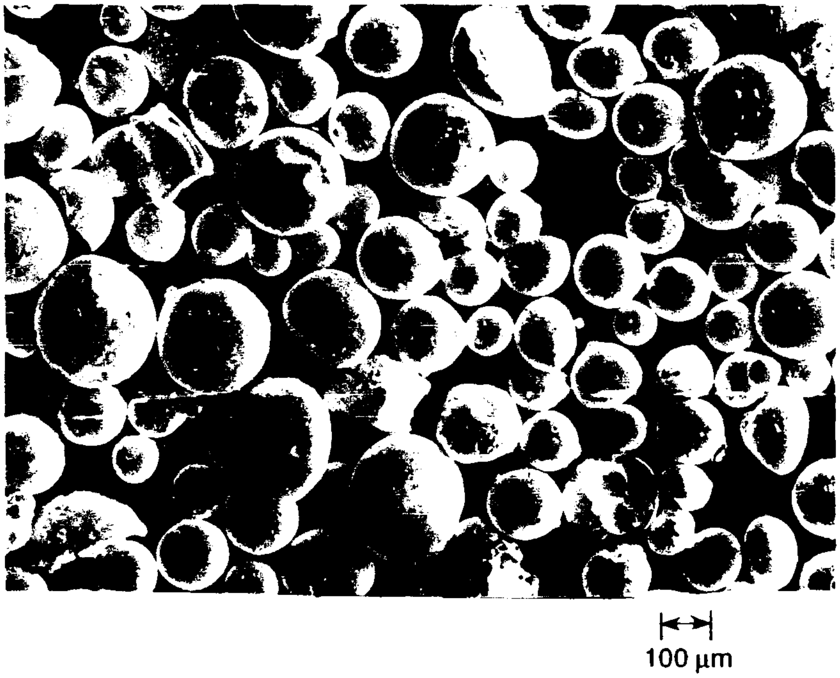
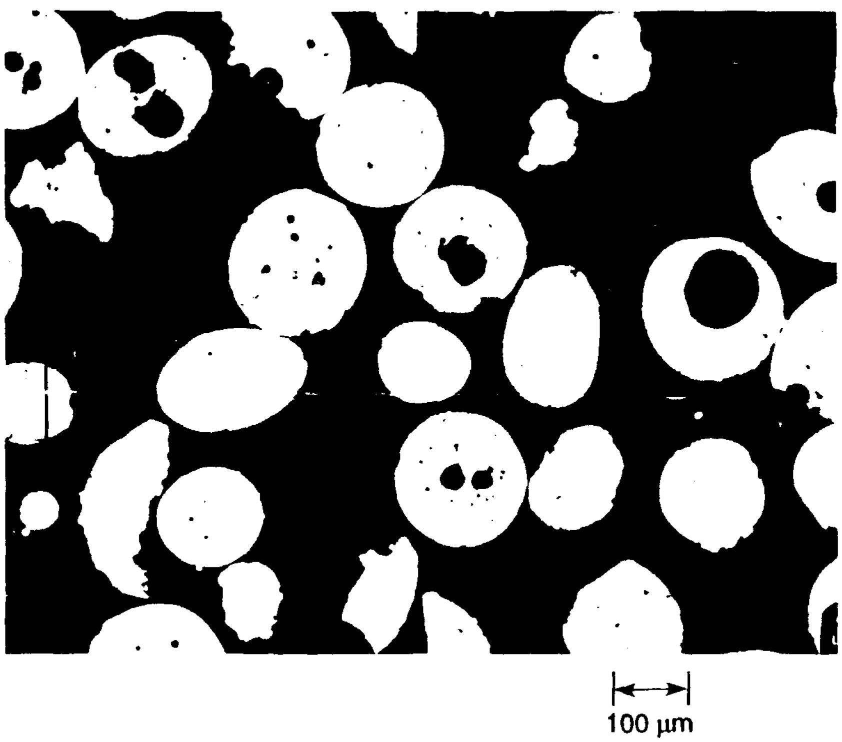


Fig. 4. Molten salt bath produced Er_3Ni; scanning electron micrograph.

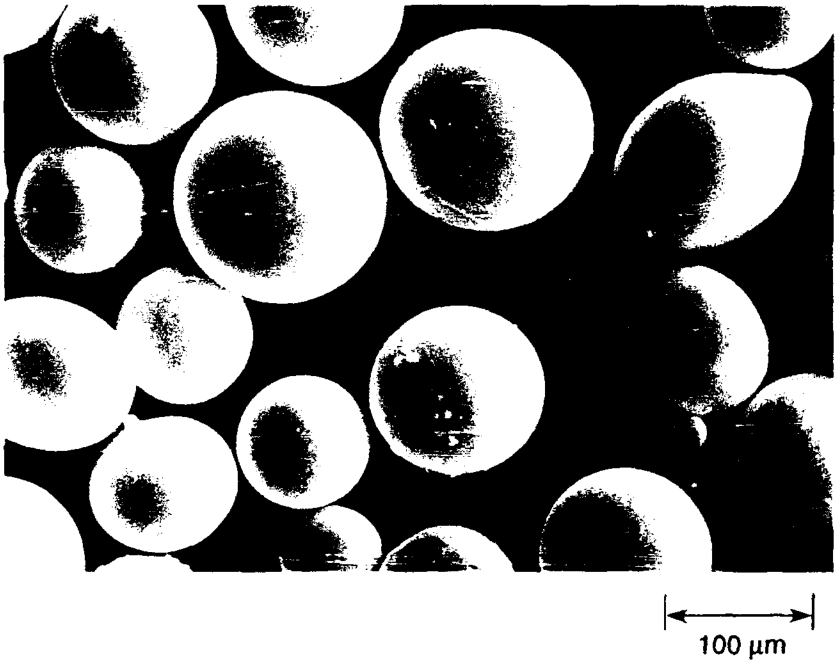


a. Scanning electron micrograph.

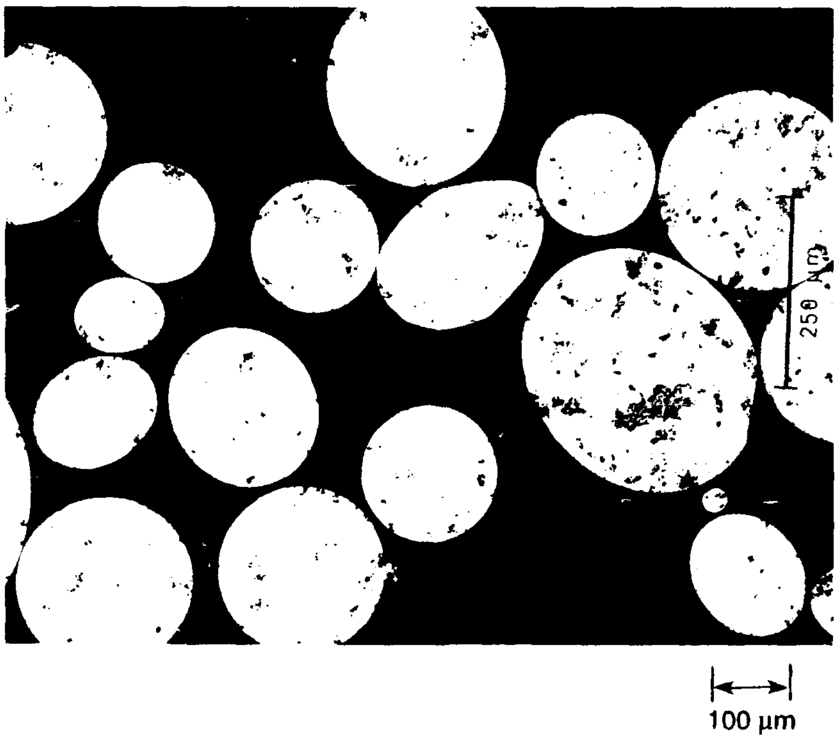


b. Optical microscope image of cross-sectioned powder.

Fig. 5. Micrographs of spray atomized Er_3Ni.



a. Scanning electron micrograph.



b. Optical microscope image of cross-sectioned powder.

Fig. 6. Micrographs of neodymium powder produced by the rotating plasma electrode method.

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